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Measurement of Acoustic Loss in Nano-Layered Coating Developed for Thermal Noise Reduction

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***Abstract*—**Structural relaxation processes in optical coatings represent a fundamental limit to the sensitivity of gravitational waves detectors, MEMS, optical metrology and entangled state experiments. To face this problem, many research lines are now active, in particular the characterization of new materials and novel solutions to be employed as coatings in future gravitational wave detectors. Nano-layered coating deposition is among the most promising techniques. We report on the measurement of acoustic loss of nm-layered composites (Ti_2O/SiO_2), performed with the GeNS nodal suspension, compared with sputtered λ/4 thin films nowadays employed.

***Keywords*—**Mechanical measurement, nanomaterials, optical coating, thermal noise.

I. INTRODUCTION

IMPORTANT advances have been made in thin-film technology during the last decade (diamond films, HTS films, superlattices and band-gap engineered films, graphene, etc.) with important fall-outs (VLSI integrated circuits, optical storage devices, photovoltaic cells, displays, micro-, nano-sensors/actuators, etc.). Optical thin-film physics and technology rely on a solid background. The study of thermal noise in thin optical films is, however, a relatively recent research theme, stemming from the needs of extreme metrology and precision measurements [1]-[3]. The present sensitivity limits to atomic spectroscopy, laser stability, atomic clock precision are set by the noise level of optical coatings. During the last few years the subject has been mainly developed in connection with thermal noise reduction in the reflective coatings of the mirrors of interferometric detectors of gravitational waves (GW) like Advanced Virgo [4] and Advanced LIGO [5].

On September 14th, 2016, the GW coming from the coalescence of a binary black hole system at cosmological distance have been detected by a coincident signal recorded by the two LIGO interferometers [6]. This first detection marks the opening of a new window toward the Universe. To allow GW astronomy be complementary to astrophysical observations in elctromagnetic spectrum, a further increment in sensitivity is required.

Interferometric detectors use the variation of travel time of light between two suspended mirrors to track the metric change induced by a passing GW. The sensitivity of Earth-bound interferometric detectors of GW is maximum in a frequency band between a few tens and a few hundred Hz, where several GW sources could be observable, and where the first GW was actually observed. The thermal noise can couple with the displacement of the mirrors of the interferometer; in particular, the Brownian noise within the high-reflectivity dielectric coatings of the test masses sets the limiting sensitivity of the nowadays advanced instruments.

The coatings used in both first- and advanced-generation GW detectors consist of alternating λ/4 layers of amorphous glassy oxides with a high and low index of refraction. According to the fluctuation-dissipation theorem [7], the spectral density of thermally activated equilibrium fluctuations is determined by the mechanical dissipative characteristics. On the basis of available evidence, dissipation in the bulk of the coating material appears to be the dominant loss mechanism, while interfacial friction between coating layers and between coating and substrate is comparatively negligible [8].

Extensive material down-selection led to the choice of SiO_2 (Silica, n=1.472) and Ta_2O_5 (Tantala, n=2.035), as the best available coating materials. The development of Ti-doped Tantala by LMA [9] was the major breakthrough in coating material engineering, and, together with the idea of coating thickness optimization [10], it was the key to the design and construction of the Advanced LIGO and Advanced Virgo coatings.

Along with the operation of advanced detectors, the GW community is currently devising a new generation of interferometers, with the aim of further enhancing the sensitivity by a factor 10. This process actually started years ago, and it encompasses today the conceptual design of third-generation observatories such as Einstein Telescope in Europe [8] and Cosmic Explorer in USA [11]. In these foreseen detectors, thermal noise will be reduced by keeping masses and suspensions at cryogenics temperatures. Unfortunately, most coating materials, including SiO_2 and Ta_2O_5, exhibit enhanced elastic losses at low temperatures, spoiling the potential benefits of mirror cooling [12].

As an alternative technology, epitaxial layers of crystalline GaAs and AlGaAs, which can be grown on a GaAs wafer and

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then transferred to a fused-silica substrate, or others crystalline materials such as AlGaP/GaP are under investigation [13], [14]. Seeking for better coating materials is not the only option. Different mirror designs, non-diffractive coating-free mirrors based on total internal reflection [15] and diffractive grating-based mirrors [16] have been also proposed, but are still in a proof-of-concept phase.

Nano-layered coatings [17], [18] can represent a promising technology at room temperature, in that they can be thermally treated to reduce the sources of losses, preventing the onset of crystallization that spoil the optical performances. Also, nm-layered composites based on materials suitable for cryogenics temperature like Titania or Hafnia are considered in view of a future, cryogenic GW detector.

II. THE NANO-LAYERED COMPOSITES

Plane-stratified glassy-oxide mixtures, where each layer is very thin (typical thicknesses are a few nm) compared to the relevant optical and elastic wavelengths, appear as effectively homogeneous as regards their optical and mechanical properties. Nano-layered composites consist of alternating nanometer-scale layers of a high index material (like TiO_2, HfO_2, or ZrO_2) and a stable low-index glass-former (e.g., SiO_2 or Al_2O_3). A homogeneous quarter wave layer with $n = 2.09$ is replaced by an optically equivalent stack of layers composed by pairs of nm-thick titania/silica layers (with titania being the first layer on top of a silicon substrate). Compared to co-sputtered glassy oxides mixtures with the same composition, nano-layered composites turn out to be optically denser, and may exhibit lower mechanical losses for the same value of the refractive index. Furthermore, the glass-former layers may effectively hinder crystallization of the high-index layers upon thermal annealing. Our measurements re aimed at testing the effectiveness and extent of these features.

A. Samples

The substrates were a set of 5 silicon disks <100>, double side polished. The disks diameter is 2 inches and their thickness 1 mm. One disk (#1) has been left bare, while the other four have been treated and/or coated (see Table I). The coating was deposited at National Tsing Hua University, Taiwan, using a 2.5cm Kaufman type ion beam sputter apparatus. The samples have been provided in the framework of the ADCOAT collaboration (funded by INFN). The deposition parameters are:1000V beam voltage, 50mA beam current and 200V accelerator voltage. A plasma bridge neutralizer was used to neutralize the ion beam. The sputtered targets were high purity Ti and SiO2 disks, 4" in diameter. They were thermally attached to either side of a water-cooled flat holder that could be flipped to face the incoming ion beam alternately, for depositing the titania and the silica layers. The coating chamber was pumped down to 10^{-6} torr for deposition. Oxygen gas was fed in the chamber with partial pressure of 10^{-4} torr for reactive sputtering. The angle of incidence of the ion beam was 45° to the target normal. The substrate holder was rotating on its axis. The surface normal of the substrate was perpendicular to the ion beam. The thickness uniformity of the coatings for both titania and silica was better than 96% within 5cm in diameter from the rotation axis. The thicknesses of the films were controlled by deposition time. Deposition rates for titania and silica films were calibrated in advance and appropriate deposition time were determined according to the design thickness.

B. Heat Treatment

The samples were thermally annealed in air. During the anneal process, the temperature was initially ramped up at a rate of 3°C/min up to the desired level and kept constant for 24 hours before the heating was turned off. The samples were then naturally cooled in the oven to room temperature. The time needed for cooling was approximately 24 hours [19]. The temperatures chosen for our samples were 250°C and 350°C (crystallization process presumably occurs at the latter temperature).

TABLE I
SAMPLES

Number #	Sample Description	Average Thickness for each layer	
		TiO_2 (nm)	SiO_2 (nm)
1	Bare Silicon wafer	-	-
2	Silicon wafer (uncoated), annealed to 250°C	-	-
3	(19-layer) Titania/Silica composite on silicon, as deposited	6.6(x10)	4.3(x10)
4	(19-layer) Titania/Silica composite on silicon, annealed to 250°C	6.4(x10)	4.3(x10)

III. MEASUREMENT OF ACOUSTIC LOSS

Loss in the material is conveniently described by the so called loss angle, that, for a resonant mode, turns to be proportional to the energy dissipated in a cycle divided by the total elastic energy. The loss angle measurements are performed exciting a resonant mode of a sample and measuring the exponential decrease of the oscillation amplitude (ring-down). Of course this implies the sample to be held by some kind of suspension or clamping and the energy transferred from the sample to the suspension to be negligible. To face this problem several solutions were adopted by different research groups involved in this kind of research, according to the various sample shapes and dimensions.

When dealing with thin slabs a massive clamp is typically used to hold the sample. Thick cylinders used as mirror masses can be suspended using a single wire that passes around the lateral surface of the cylinder, forming a "U" shaped suspension, narrow in the upper part [20]. Furthermore, if the specimen is fused silica, it is possible to weld a thin silica fiber directly on the edge of the sample [21]. Cylindrical thin samples can be clamped between two spheres, pressing against the center of the two faces [22]. In this configuration, the transferred energy for those modes having the centers as nodal points is negligible. In all these systems the experimentalists do not easily know whether the measured loss angle is a property related to the material or sample under investigation or it is limited by the excess loss coming from their setup. The problem is relevant when ultra-low loss materials are under investigation. Parameters such as the effective contact surface, the specific damping due to the contact between two materials, and even the precise location of the contact surface, are difficult to quantify and to control.

A. *Experimental Set-up GeNS*

The GeNS suspension does not present any intrinsic limitation to high Q measurements, when comparing the results with the ones from other laboratories that measured similar samples with the same aspect ratio. In this solution the sample, which for our purposes is disk shaped, is suspended in equilibrium on top of a sphere or half-sphere. In this kind of suspension, the contact surface is minimized because of the absence of applied forces. It is suitable for studying the mechanical dissipation of coated samples, because it leaves one surface untouched and does not damage the film on it. Moreover, it allows us to change one at a time the different relevant parameters of the set up that may affect the measurements, distinguishing in principle the intrinsic dissipation of the material from the losses due to the experimental setup (see Fig. 1). If θ is the angular position of the disk, suspended on the sphere, with respect to the horizontal plane, a stable equilibrium is achieved whenever $D > t$, where D is the diameter of the sphere and t is the thickness of the suspended cylindrical sample. For a pure rolling condition, the angular range where stable oscillation is possible is:

$$\vartheta \approx \sqrt{(3(\mathrm{D} - \mathrm{t})/\mathrm{t})} \tag{1}$$

Usually the stability region is large enough to guarantee a fast and handy manual positioning. The GeNS experimental set up and its main features have been fully described in a previous article [23].

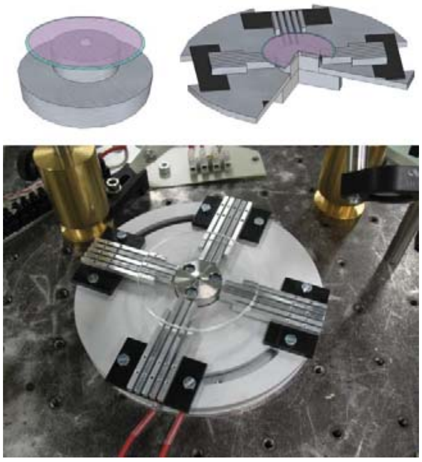

Fig. 1 A schematic view and a picture of the GeNS set up, highlighting the sphere holder and the four comb exciters

B. *Loss Angle Measurements*

We looked at the decay time of the free oscillation at 5 resonance frequency of each disk. This set of modes only includes vibrations with radial nodal lines (butterfly modes, see Fig. 2).

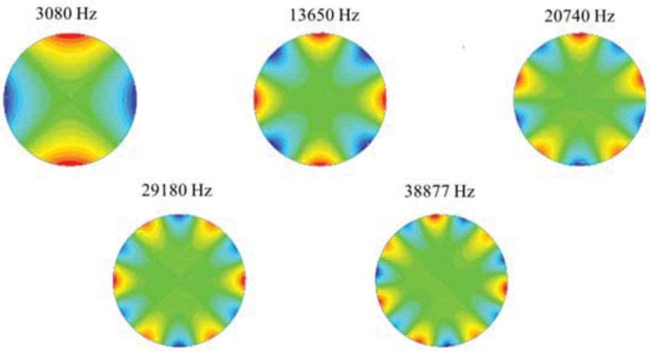

Fig. 2 A view of the resonance butterfly mode simulated with finite element analysis (ANSYS), corresponding to resonance frequency measured

The loss angle is then worked out from the decay time τ, taking into account the mode frequency f, as

$$\phi = \frac{1}{\pi f \tau} \tag{2}$$

The oscillation is read out with an optical lever. The excitation is provided through four comb exciter placed under the sample and the optical lever displacement is read by a shadow meter photodetector. The results for samples #1, #2, #3 and #4 are reported in Fig. 3. We assume for each measurement a relative error of 5%, estimated as the reproducibility for different suspensions.

Fig. 3 also shows a comparison with the disk estimated thermoelastic damping [24], which, for our disks, is expected to be the main contribution to the substrate losses. Evidence is collected, that the annealing of sample #2 removed a spurious source of loss, bringing the overall loss angle to the level of thermoelastic dissipation. The origin of such spurious loss is still not clear, but can be traced to machining procedures or contaminants.

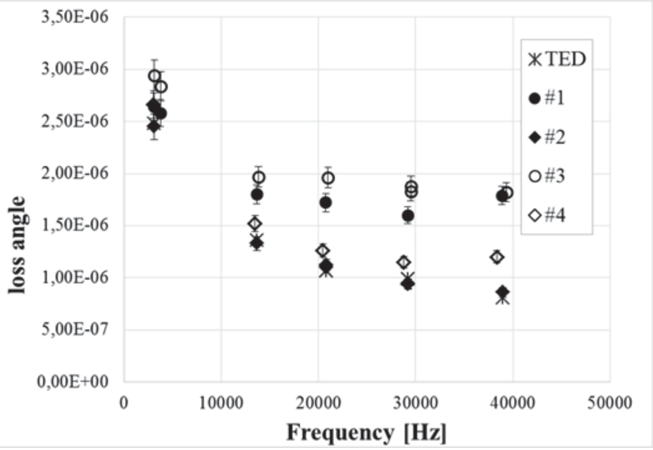

Fig. 3 Loss angle measured with GeNS, for disk samples #1, #2 and #3, for 5 resonant modes

IV. COATING LOSS EXTRACTION

The coating loss angle can be roughly estimated [25] from the difference $\Delta\phi$ between the measurement of the coated sample and the measurement of bare sample using an approximate formula for thin film. In particular, in our case we compute the difference between sample #3 and samples #1, and the difference between sample #4 and sample #2 using:

$$\phi_{layer} = \frac{Y_S t_S}{3(Y_{Ti} t_{Ti} + Y_{Si} t_{Si})} \Delta\phi \qquad (3)$$

where Y and t are the Young's modulus and the thickness of substrate (s), of TiO_2 (Ti) and of SiO_2 (Si) nanolayers.

Assuming Y_{Ti} = 165 GPa, Ysi =72 GPa, Y_s= 150 GPa (effective value for a plate bending in <100> silicon) we obtained the loss angles for nano-laminated stack coating as shown in Fig. 4. Supposing the coating loss angle approximatively independent from the frequency, and excluding the points at highest frequency, probably affected by resolution limits of the detection system, we obtained for the nano-layered composite a value of $\varphi_{nc} = (8.7 \pm 1.7) \cdot 10^{-4}$, and for the annealed one $\varphi_{nc} = (7.2 \pm 0.8) \cdot 10^{-4}$.

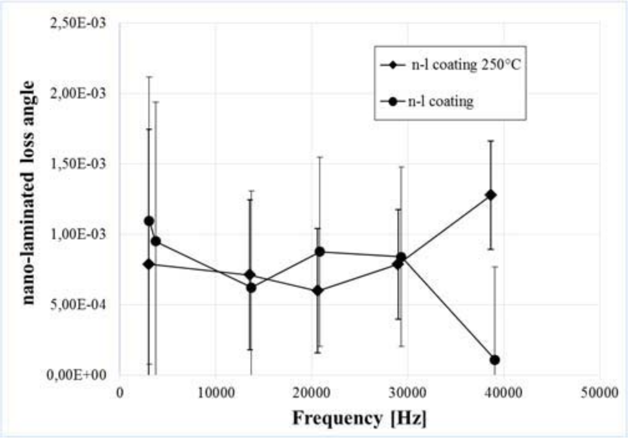

Fig. 4 Calculated loss angle for 19 nano-layers for 5 resonance modes for #3 and #4 sample

V. CONCLUSION

The results show a loss angle of the same order of the one measured by other system, but the effect of annealing is not so evident [26]. However, our measurements have a better confidence range.

The previous measurements have been done on substrates with different shapes (silicon micro-cantilevers), that have lower resonance frequencies and smaller area of coating. This last point could be determinant. This is the first time that the NTHU coater set up deposited the nano-laminated stack layer on an area of 2 inches of diameter. Some hints could be provided by the measure of the acoustic attenuation of samples with a nano-laminated coating deposited on a smaller area (diameter of 1 inches). Another important issue is the bare sample preparation: Starting with substrates with no additional losses to thermoelastic damping, or annealing always the substrates before the coating deposition, or changing silicon wafers with other with low resistivity, low impurity level, and good mechanical performances could be possible ways forward. However, while the mechanical performances are comparable with sputtered λ/4 thin films nowadays employed, nano-laminated stack coating still have the possibility of higher annealing without increasing the crystallite dimensions. An upper layer of SiO_2 is recommended to preserve the surface and to block the crystallite growth in the external layer.

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E. Cesarini (Urbino, May 14th 1979) is a physicist, member of the LIGO-Virgo collaboration since 2006. She graduated at Università degli studi di Bologna (IT) in 2004 with a thesis in astrophysics. Then she did the phd at Università degli studi di Firenze (IT) and she discussed her thesis on mirror thermal noise in gravitational wave detectors in 2009. Then she spent some years as a post doc in Florence (IT), a semester at University of Texas, Brownsville (US) with a research scholarship and finally she reached the Virgo group at Università di Roma Tor Vergata (IT) in 2012. She has been involved in many topics on thermal noise in present and future detectors of gravitational wave, becoming expert in particular in acoustic loss measurement of coating. She is also inside the ADCOAT collaboration (funded by INFN). Furthermore, she is involved in thermal compensation aberration in interferometric detectors of gravitational waves. The encountered issues in her career bring her to enter in contact with many field regarding science materials. It has some hundreds of publications inside the scientific collaboration and ten few names publications. She has been awarded as best student poster in 2009 at 8th Amaldi Conference in New York (US).

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